

## PATENT ABSTRACTS OF JAPAN

(11)Publication number : 11-343152

(43)Date of publication of application : 14.12.1999

(51)Int.Cl.

C03C 27/12

(21)Application number : 10-203425

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(22)Date of filing : 17.07.1998

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(30)Priority

Priority number : 09192474	Priority date : 17.07.1997	Priority country : JP
09213284	07.08.1997	JP
09223613	20.08.1997	JP
09247015	11.09.1997	JP
09253216	18.09.1997	JP
10 983	06.01.1998	JP
10 22137	03.02.1998	JP
10 91947	03.04.1998	JP

(54) INTERMEDIATE FILM FOR LAMINATED GLASS AND LAMINATED GLASS

(57)Abstract:

PROBLEM TO BE SOLVED: To obtain a laminated glass which maintains basic performance necessary for a laminated glass and little efflorescence in the peripheral part even in a high humidity atmosphere by forming an intermediate film comprising a plasticized polyvinyl acetal resin film having a specified or lower haze when the film is immersed in water.

SOLUTION: When the intermediate film for a laminated glass comprising a plasticized polyvinylacetal resin film having 0.3 to 0.8 mm thickness is immersed in water at 23° C, efflorescence of the film after 24 hours is controlled to  $\leq 50$  % haze. To obtain the haze value above described, sodium salts and potassium salts which cause the efflorescence and which are mixed from the source material are controlled to have  $\leq 10$   $\mu$ m particle size and to give  $\leq 50$  ppm sodium concn. and  $\leq 100$  ppm potassium concn. in the intermediate film. To effectively prevent efflorescence at high humidity, compds. which can form complexes with sodium salts and potassium salts, and org. acids and amines compatible with the resin and the plasticizer are preferably added as dispersants.

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## TAILED DESCRIPTION

tailed Description of the Invention]

[01]

id of the Invention.] This invention relates to the glass laminate which used the interlayer for glass  
inates, and the above-mentioned interlayer for glass laminates.

[02]

scription of the Prior Art] The glass laminate with which it comes conventionally to fasten the  
rayer which consists of a plasticized polyvinyl butyral between the glass plates of at least two  
ets, it has basic performance transparency, weatherability, and an adhesive property are good,  
d penetration resistance moreover and required for the glass laminate of a glass fragment not  
ersing easily, for example, is widely used for the winduppane of a car or a building.

[03] This kind of glass laminate is inferior in moisture resistance, although the above-mentioned  
ic performance is good and excellent in safety. That is, by the periphery of a glass laminate, when  
above-mentioned glass laminate is placed into a humid atmosphere, since the interlayer touches  
ronmental air directly, the problem which the interlayer of a periphery milks arises.

[04] The additive agent for performing adhesive strength adjustment of an interlayer and glass  
cribed below is participating in this abtism. In order to demonstrate the function as the above-  
ioned glass laminate enough, it is required to adjust the adhesive strength of an interlayer and  
is so that it may fall within a proper range. Namely, if the danger of the glass fragment damaged by  
shock from the outside, etc. separating and dispersing from an interlayer, and doing injury to a  
nan body etc. if the adhesive strength of an interlayer and glass is too weak becomes high and the  
esive strength of an interlayer and glass is too strong conversely. Glass and an interlayer are  
utaneously damaged by the shock from the outside, etc., and the danger of the adhesion fragment  
glass and an interlayer dispersing and doing an obstacle to a human body etc. becomes high.

[05] On the other hand, when there is adhesive strength of an interlayer and glass within proper  
ts. Since the phenomenon in which partial interfacial peeling of an interlayer and glass happens,  
an interlayer extends arises at the same time glass is damaged while glass breakage reaches far  
l wide and happens, the impact-absorbing effect and a penetration preventive effect become large.

[06] Therefore, in case of the accident of transport-airplane machines, such as a car, in order to  
ort a shock in case a driver and a passenger collide to glass or to prevent penetration, in case of  
accident of a building, in order to prevent the missile from the outside from penetrating glass or  
prevent scattering of a glass fragment, it is required to adjust the adhesive strength of an  
rayer and glass so that it may fall within a range proper like \*\*\*.

[07] In order to adjust the adhesive strength of an interlayer and glass within proper limits  
entionally in view of the above, the adhesive strength regulator for interlayers has been

mined variously. In JP 46-4270.B, the interlayer for glass laminates which consists of a polyvinyl-  
tal resin composition which carries out specific amount content of 0.2 to 0.8 % of the weight of  
sture and the metal alkyl carboxylate specific as an adhesive strength regulator is proposed. The  
rayer by the above-mentioned proposal tends to adjust the adhesive strength of an interlayer  
i glass to the proper range by changing the distribution quantity of metal alkyl carboxylate in an  
rayer layer part and an interlayer inner layer part, or changing the moisture content in an  
rayer.

[08] However, the interlayer containing metal alkyl carboxylate like the above-mentioned proposal,

Since the interlayer is carrying out direct contact to air in the periphery of the glass laminate if the  
glass laminate which moisture resistance fell and was manufactured using this interlayer is neglected  
under a humid atmosphere. While the quantity of metal alkyl carboxylate increases, there is a problem  
that the abtism by moisture absorption of an interlayer happens violently. Although it can prevent  
whether the abtism of the above-mentioned interlayer reduces the quantity of metal alkyl  
carboxylate as much as possible, and by losing, in that case, the fatal problem as a glass laminate of  
the adhesive strength of an interlayer and glass becoming stronger than the proper range too much,  
and glass and an interlayer being simultaneously damaged by the shock from the outside, etc., or  
becoming easy to penetrate had occurred.

[0009] In JP 44-32185.B, contain 0.1 to 0.8% of moisture, and The monocarboxylic acid of six to 22  
carbon atom. The dicarboxylic acid of four to 12 carbon atom, and the aliphatic series monosamin  
monocarboxylic acid of two to 6 carbon atom. The interlayer for glass laminates which carries out  
0.01-3 weight-section content of the aliphatic series monosamin dicarboxylic acid of four to 5 carbon  
atom and at least one organic acid chosen from citrate and these mixtures per resin 100 weight  
section and which consists of shaping polyvinyl-acetal resin is proposed.

[0010] However, when carboxylic acid is added, there is a problem that adhesive strength changes  
with time. The problem that the heat resistance and the weatherability of an interlayer fall under the  
influence of acid arises.

[0011] In the glass which pasted the glass of at least two sheets together with the plasticized-  
polyvinyl-acetal-resin constituent in JP 48-6772.B, The glass laminate making sodium metal salt of  
the aliphatic carboxylic acid of the carbon numbers 10-22 contain in this plasticized-polyvinyl-  
acetal-resin constituent is proposed.

[0012] In JP 53-18207.B, using the alkali metal salt or alkaline earth metal salt of monocarboxylic acid  
or dicarboxylic acid is proposed as an adhesive strength regulator in the plasticized-polyvinyl-acetal-  
resin interlayer of a glass laminate.

[0013] In the two above-mentioned proposals, each uses metal salt of carboxylic acid with a  
comparatively large carbon number as an adhesive strength regulator from it being easy to dissolve in  
the plasticizer contained in an interlayer.

[0014] However, when metal salt of carboxylic acid with a large carbon number is used as an adhesive  
strength regulator, there is a problem that the adhesive strength of an interlayer and glass changes  
with time progress (temporality). That is, even if early adhesive strength is proper, adhesive strength  
declines gradually with temporality, and when shocked, glass exfoliates easily. In order to prevent this  
adhesive strength fall, it is necessary to keep an interlayer for one to two months under 40-50 °C  
atmosphere, and to ripe, and. Since an interlayer has adhesiveness, autohesion nature, etc., even if  
keeping it under the above atmosphere for a long period of time rines difficult actually, although the  
fall of adhesive strength with the passage of time can be controlled, it cannot be made for there to  
be nothing, but the above-mentioned problem will still remain.

[0015] In JP 60-210551.A, to plasticized-polyvinyl-acetal-resin 100 weight section. The glass laminate  
with which the glass of at least two sheets sticks, and it comes to unite it by whether 0.02 to  
potassium monocarboxylate 0.40 weight section and 0.01 to modified silicone oil 0.20 weight section  
whose carbon numbers are 1-4 contain, and the interlayer to which it adheres is indicated. However,  
since this metal salt solidified in particle state in the interlayer depending on the kind of metal salt  
used and this glass laminate caused a white bluish mark, the perfect thing was not able to say from a  
viewpoint of preventing the white bluish mark by long-term moisture absorption.

[0016] In JP 2-41547.B, the polyvinyl-butyl sheet which uses alkali or alkaline-earth-metal formate  
for an adhesive strength regulator is proposed. In the Patent Publication Heisei No. 502594 [ six to ]  
gazette, the interlayer which added potassium acetate as an adhesive strength regulator in the  
example is used.

[0017] In the three above-mentioned proposals, in order to cancel acid problem in the case of using  
metal salt of carboxylic acid with a large carbon number as an adhesive strength regulator, metal salt  
of carboxylic acid with a comparatively small carbon number is used

[0018] However, if metal salt of carboxylic acid with a small carbon number is used as an adhesive  
strength regulator. Although the problem of an adhesive strength fall with the passage of time of an  
interlayer and glass is canceled, the moisture resistance of an interlayer becomes insufficient and, as  
a result, another problem of becoming easy to start the abtism by moisture absorption to the edge

t (end) of a glass laminate occurs.

19) That is, since hygroscopicity is under the usual atmosphere (humidity), when processing it into glass laminate, it is common [an interlayer] to process it by controlling the humidity and doubling, example so that water content may be about 0.5 or less % of the weight under the atmosphere of RH. However, since the edge part of a glass laminate is in an unreserved state, under a high humidity atmosphere, an interlayer absorbs moisture and water content usually rises to about 2 to 3% weight. At this time, water gathers for the circumference of metal salt of carboxylic acid with a carbon number of potassium acetate, magnesium acetate or potassium formate etc, which acts as a minute crystal in an interlayer, and albinism is caused. If the addition of carboxylic acid in a small carbon number or its salt is decreased in order to reduce albinism, the adhesive strength in interlayer and glass will deviate from the proper range, and will become insufficient [impact-option nature, penetration resistance, etc. of a glass laminate].

20) As a trial to improve, the white bluish mark of the interlayer containing carboxylic acid metal salt, in JP 5-188250A. The interlayer for glass laminates currently formed from polyvinyl-acetal resin, plasticizer, and the resin composition in which aliphatic series mono- or di-carboxylic acid is or less contains the alkali or alkaline earth metal salt, and organic acid of dicarboxylic acid is posed.

21) The interlayer for glass laminates which becomes JP 7-41340A from the resin composition containing polyvinyl-acetal resin, a plasticizer, carboxylic acid metal salt, and straight-chain fatty acid composed.

22) However, the glass laminate using the interlayer for glass laminates of the above-mentioned proposal is not still enough, although the white bluish mark of the edge part after a humidity resistance test is reduced. And when it is going to reduce a white bluish mark further and the content of straight-chain fatty acid is increased, and a glass laminate is placed under relatively high temperature, there is a possibility of producing foaming and discoloration.

23) Although the interlayer of the above-mentioned proposal tries solution of a white bluish mark improvement of an adhesive strength regulator, also in the interlayer which is not adding the adhesive strength regulator, the white bluish mark by moisture absorption produces it. It became clear our latest research that the inquiry in the resin described below is involving as one of the cause this.

24) Although the interlayer for glass laminates of this invention uses polyvinyl-acetal resin as the ingredients, the neutralization process is included when manufacturing polyvinyl-acetal resin. In the solution of sodium salt, such as sodium hydroxide and sodium bicarbonate, is used in this neutralization process, for example. When those sodium salt is used superfluously and new sodium acetate by neutralization, sodium salt remains in the polyvinyl-acetal resin obtained.

A sodium salt that remained causes [big] a white bluish mark by moisture absorption of the interlayer for glass laminates obtained in order to promote condensation of water, when it becomes a solid state at the time of a polymerization and desiccation and polyvinyl-acetal resin absorbs water. Sodium salt may remain also to polyvinyl alcohol and this sodium salt may cause a white bluish mark by moisture absorption of the interlayer for glass laminates.

25) In recent years, the motion which uses a glass laminate for side glasses and the various kinds of car posters, in these uses, also when using the periphery of a glass laminate in the reserved state, it is increasing, and the request to albinism prevention is increasingly stronger.

26) The object(s) to be Solved by the Invention [The place which this invention solves the above-mentioned problem and is made into the purpose, Without spoiling basic performance required for glass laminates, such as transparency, weatherability, an adhesive property, and penetration resistance, even when moreover placed into a humid atmosphere, there is a white bluish mark of a glass laminate edge part in providing the interlayer for [little] glass laminates, and the glass laminate is it.

27) The means for Solving the Problem [This invention is an interlayer for glass laminates which consists of plasticized polyvinyl polyvinylacetal resin films, and when the 0.3-0.8-mm-thick above-mentioned interlayer is immersed in 23 ° water, Hayes of 24 hours after is an interlayer for glass laminates which is 50% or less. This invention is explained in full detail below.

[0028] When an interlayer for glass laminates of this invention immerses a 0.3-0.8-mm-thick interlayer in 23 ° water, Hayes of 24 hours after is 50% or less.

[0029] When this invention persons immerse a 0.3-0.8-mm-thick interlayer in 23 ° water. Even when Hayes of 24 hours after was placed into atmosphere where 50% or less of interlayer for glass laminates is humid, there were few white bluish marks of a glass laminate edge part, they found out excellent in moisture resistance, and completed this invention.

[0030] Since it becomes insufficient to prevent a white bluish mark under high humidity and it is inferior to moisture resistance when above-mentioned Hayes exceeds 50%, it is limited to a mentioned range. Above-mentioned Hayes as used herein shall mean a value which used and measured an integral equation turbidity meter 24 hours afterward, when a 0.3-0.8-mm-thick interlayer is immersed in 23 ° water.

[0031] An interlayer for glass laminates of this invention consists of a plasticity polyvinyl-acetal resin layer, and the above-mentioned plasticity polyvinyl-acetal resin layer uses polyvinyl-acetal resin as the main ingredients.

[0032] As the above-mentioned polyvinyl-acetal resin, average degree-of-acetalization% of a thing of 40-75 mol is preferred, compatibility with a plasticizer falling that it is less than [40 mol %], and mixing a plasticizer of a complement to reservation of penetration resistance --- hard --- --- there is a case, if 75-mol % is exceeded, while a mechanical strength of an interlayer for glass laminates obtained will fall, in order to obtain resin, prolonged reaction time is required, and it is not sometimes desirable on a process. It is 60-75-mol % more preferably, if it is less than [60 mol %], since hygroscopicity becomes high, a white bluish mark will happen easily. It is 64-71-mol % preferably.

[0033] In the above-mentioned plasticized polyvinyl-acetal resin, the following [30 mol %] have a preferred vinyl acetate component. Since it will become easy to cause blocking at the time of manufacture of resin if 30-mol % is exceeded, it becomes difficult to manufacture. Preferably, it is less than 19 mol %.

[0034] The above-mentioned plasticized polyvinyl-acetal resin comprises a vinyl-acetal ingredient, a vinyl alcohol ingredient, and a vinyl acetate component, and each of these component amounts. It can measure based on, for example, a JIS K 6728 "polyvinyl-butyl test method" and a nuclear magnetic resonance method (NMR).

[0035] When the above-mentioned polyvinyl-acetal resin is except polyvinyl butyral resin, a vinyl alcohol component amount and the amount of vinyl acetate components can be measured, and the remaining vinyl-acetal component amounts can be computed by deducting both the above-mentioned component amounts from 100.

[0036] The above-mentioned polyvinyl-acetal resin can be conventionally manufactured by a publicly known method. For example, solution obtained by dissolving polyvinyl alcohol in warm water --- a predetermined temperature --- for example, it holds at 10-20 ° preferably, and 0-95 % of acetalization reactions are advanced, adding and stirring a necessary acid catalyst and aldehyde. Subsequently, reaction temperature is raised to 70 °, it ripens, a reaction is completed, and a method of performing neutralization, rinsing, and desiccation and obtaining powder of polyvinyl-acetal resin, etc. are mentioned after that.

[0037] As polyvinyl alcohol used as the above-mentioned raw material, a thing of the average degrees of polymerization 500-5000 is preferred, and a thing of the average degrees of polymerization 1000-2500 is more preferred. When it is less than 500, the penetration resistance of a glass laminate obtained may fall, if 5000 is exceeded, shaping of a resin layer will carry out --- hard --- --- there are things and, moreover, intensity of a resin layer may become strong too much.

[0038] Since it is preferred to set a vinyl acetate component of polyvinyl-acetal resin obtained to less than 30 mol %, therefore, a saponification degree of the above-mentioned polyvinyl alcohol has a preferred thing beyond 70 mol %. When it is less than [70 mol %], the transparency of resin and heat resistance may fall and reactivity may also fall. It is a thing beyond 95 mol % more preferably.

[0039] An average degree of polymerization and a saponification degree of the above-mentioned polyvinyl alcohol can be measured, for example based on JIS K 6728 "polyvinyl alcohol test-method." As the above-mentioned aldehyde, aldehyde of the carbon numbers 3-10 is preferred. The moldability of resin layers with a carbon number enough by less than three may not be obtained. If 10 is exceeded, the reactivity of acetalization will fall, and moreover it will become easy to generate a block of resin during a reaction, and will become easy to follow difficulty on composition of resin.









(111)

81][R<sup>5</sup>, R<sup>6</sup>, and R<sup>7</sup> are a hydrogen atom, an aliphatic series system hydrocarbon group whose ben numbers are 1-20, or an aromatic system hydrocarbon group whose carbon numbers are 1-20 and a formula, respectively.) R<sup>5</sup>, R<sup>6</sup>, and R<sup>7</sup> may be the same, and may differ from each other.

82]When a carbon number of the above-mentioned aliphatic series system hydrocarbon group or above-mentioned aromatic system hydrocarbon group exceeds 20, it may become hydrophobicity compatibility with polyvinyl-acetal resin may worsen. As for any one of R<sup>5</sup>, R<sup>6</sup>, and R<sup>7</sup>, it is preferred that it is a long chain, and a hydrocarbon group and R<sup>7</sup> R<sup>5</sup> and whose R<sup>6</sup> are a hydrogen atom and the carbon numbers 1-2 are a hydrocarbon group of the carbon numbers 6-16 more preferably.

83]As amine expressed with the above-mentioned general formula (III). For example, they are defined by nitrogen-containing heterocyclic compounds, such as aromatic amines, such as 1, 2-alkylamine of the 3rd class, and aniline, and pyridine, etc., and specifically, Methylamine, ethylamine, propylamine, hexylamine, octylamine, decyl amine, dodecyl amine, tetradecylamine, and octadecylamine. The 1st class amine, such as naphthylamine; Dimethylamine; diethylamine; Secondary amine, such as dipropyl amine, diethyl amine, dioctyl amine, and N-methylamine; Trimethylamine; tertiary amine, such as triethylamine, N,N-dimethylhexylamine, N,N-dimethyl octylamine, N,N-ethyldecyl amine, N,N-dimethyldodecyl amine, N,N-dimethyleniline, and pyridine, etc. are mentioned. These may be used alone and may use two or more sorts together.

84]As for an addition of amine expressed with the above-mentioned general formula (III), when a ben number uses sulfonic acid which is 2-21 as organic acid which is compatible in resin and a sticizer, it is preferred that they are 0.01 - the amount part of duplex to polyvinyl-acetal resin weight section. When a preventive effect of a white bluish mark by moisture absorption may become insufficient when it is less than 0.01 weight section, and the amount part of duplex is needed, compatibility with resin becomes poor, a problem may arise in transparency or an rlayer may color it. It is 0.02 to 1 weight section more preferably.

85]As for an addition of amine expressed with general formula (III), when a carbon number uses boxylic acid which is 2-20 as organic acid which is compatible in the above-mentioned resin and a sticizer, it is preferred that it is 0.01 to 3 weight section to polyvinyl-acetal resin 100 weight section. When a preventive effect of a white bluish mark by moisture absorption may become inefficient if it is less than 0.01 weight sections and three weight sections are exceeded, compatibility with resin becomes poor, a problem may arise in transparency or an interlayer may color it. It is 0.05 to 1 weight section more preferably.

86]As for an addition of amine expressed with general formula (III), when using phosphoric acid ressed with the above-mentioned general formula (II) as organic acid which is compatible in the ve-mentioned resin and a plasticizer, it is preferred that they are 0.01 - the amount part of lexs to polyvinyl-acetal resin 100 weight section. When a preventive effect of a white bluish mark moisture absorption may become insufficient when it is less than 0.01 weight sections, and the ount part of duplex is exceeded, compatibility with resin becomes poor, a problem may arise in ransparency or an interlayer may color it. It is 0.05 to 1 weight section more preferably.

87]As for the above-mentioned organic acid or amine, it is preferred to choose molecular ture and a molecular weight suitably and to be used according to the degree of acetalization of above-mentioned polyvinyl-acetal resin or a kind of the above-mentioned plasticizer.

88]Amine which is compatible in organic acid, resin, and a plasticizer which are compatible in resin a plasticizer which are used as the above-mentioned dispersing agent. Respectively it becomes such as sulfonyl ion, a carboxyl ion, phosphoric acid ion, and AMMONIUM ion, it acts on the face of metal salt of particle state which exists in polyvinyl-acetal resin, and combines with a tal ion which constitutes this metal salt, and its counter ion, or [ and / that metal salt which these combined by kneading resin at the time of film production is distributed in resin, and metal salt of tic state becomes small as the result. ] --- or it disappears. For this reason, even if local densation of water is controlled and polyvinyl-acetal resin absorbs moisture, a white bluish mark

of an interlayer for glass laminates obtained can be controlled. Among organic acid which is compatible in resin and a plasticizer which are used as the above-mentioned dispersing agent, especially in phosphoric acid, since a role of a coupling agent of resin and glass is also played, and it has the character in which a film and glass do not exfoliate easily even if it absorbs moisture, exfoliation by moisture absorption of a glass laminate periphery can be controlled.

89]As for an interlayer for glass laminates, in this invention, it is preferred to contain at least one sort chosen from a group which consists of alkali metal salt and alkaline earth metal salt as an adhesive strength regulator.

90]It is not limited especially as the above-mentioned alkali metal salt and alkaline earth metal salt, for example, salts, such as potassium, sodium, and magnesium, are mentioned. As the above-mentioned salt, inorganic acid, such as organic acid; chlorides, such as carboxylic acid, such as acetic acid, hexylacid, butanoic acid, acetic acid, and formic acid, and nitric acid, is mentioned.

91]As the above-mentioned alkali metal salt and alkaline earth metal salt, it is more preferred that they are alkali metal salt of organic acid of the carbon numbers 5-16 and the alkaline earth metal salt of organic acid of the carbon numbers 5-16. It is magnesium salt of carboxylic acid of the carbon numbers 6-10, or dicarboxylic acid preferably.

92]It is not limited especially as magnesium salt of the above-mentioned carboxylic acid or dicarboxylic acid. For example, 2-ethylbutanoic acid magnesium, valeric-acid magnesium, hexanoic acid magnesium, magnesium heptanoate, octanoic acid magnesium, magnesium nonanoate, decanoic acid magnesium, glutaric acid magnesium, adipic acid magnesium, etc. are mentioned.

93]Magnesium salt of carboxylic acid of the above-mentioned carbon numbers 6-10, or dicarboxylic acid, by existing in a form of a salt, without ionizing in a film, and drawing a water molecule near, it is thought that it is possible to stop adhesive strength between an interlayer and glass, and penetration-proof performance of a glass laminate obtained by this can be made good. Since it is distributed over a membrane surface at high concentration, without condensing in a film and an excessive white bluish mark at the time of moisture absorption is not caused while a small quantity shows an adhesive strength adjustment effect, it is desirable.

94]As for the above-mentioned alkali metal salt and alkaline earth metal salt, it is preferred that particle diameter is 3 micrometers or less, and they are 1 micrometer or less more preferably. Since it will grow up to be even a size by a water molecule which gathered around alkali metal salt and/or alkaline earth metal salt is visualized if it exceeds 3 micrometers, a white bluish mark may become remarkable and may not be preferred.

95]It is not limited especially as a means for the above-mentioned particle diameter to be 3 micrometers or less. For example, even if it is a method of using a compound which is easy to dissolve in polyvinyl-acetal resin or a plasticizer as an adhesive strength regulator, and a compound which is hard to dissolve in polyvinyl-acetal resin or a plasticizer, A method of using what it is hard to condense in polyvinyl-acetal resin and a plasticizer, a method of using together a dispersing agent which distributes them, a compatibilizer, etc., etc. are mentioned.

96]As a compound which is easy to dissolve in the above-mentioned combination, polyvinyl butyral resin is used as polyvinyl-acetal resin, for example, in combination using triethylene glycol 2-ethyl butyrate as a plasticizer, organic acid salt, such as octanoic acid magnesium, neo decanoic acid magnesium, and adipic acid magnesium, etc. are mentioned, and these one sort or two sorts or more are used suitably.

97]As a potassium compound which is easy to dissolve in the above-mentioned combination, potassium salt of organic acid, such as octanoic acid potassium, neo decanoic acid potassium, and potassium stearate, etc. are mentioned, and these one sort or two sorts or more are used suitably. 98]As a sodium compound which is easy to dissolve in the above-mentioned combination, sodium salt of organic acid, such as sodium octanoate, neo decanoic acid, and sodium stearate, etc. are mentioned, and these one sort or two sorts or more are used suitably.

99]As a compound which is during combination of what it is hard to dissolve in the above-mentioned combination, and is hard to condense, magnesium salt of inorganic acid, such as a magnesium chloride and a magnesium nitrate, etc. are mentioned, and these one sort or two sorts or more are used suitably, for example.

100]As a dispersing agent which may be distributed according to concomitant use, or a compatibilizer, a compound which is hard to dissolve in the above-mentioned combination. Although

limited in particular, long chain system organic acid, such as alcohols and octanoic acid, such as amol and octyl alcohol, and nonanoic acid, is mentioned, and these one sort or two sorts or more used suitably, for example.

[01A] Method of using a compound which itself tends to dissolve in polyvinyl-acetal resin or a sticizer also in various above-mentioned methods is the most preferred, and a method of using a round which it ranks second and is hard to condense in polyvinyl-acetal resin and a plasticizer is ferred.

[02]As the above-mentioned alkali metal salt and alkaline earth metal salt, when using a diester tem compound as a plasticizer, it is preferred that it is what has an acid component and the cture of a diester system compound. Since it can distribute stably and uniformly and can exist in in by having a structure similar to an acid component of a diester system compound used as a sticizer, aging is not caused.

[03]As the above-mentioned plasticizer, triethylene glycol di-2-ethyl butyrate, When using (it also is hereafter called "3GH") or a diethyl horse mackerel peat (henceforth "DHA"), by making metal ; of the carbon number 5 or carboxylic acid of 6 contain as an adhesive strength regulator. Since a of adhesive strength of an interlayer and glass with the passage of time can be prevented and vention from a white bluish mark and prevention from a fall with the passage of time of adhesive ngth can be reconciled, it is desirable. When using triethylene glycol di-2-ethylhexanoate neforth "3GO") as a plasticizer for same reason, it is preferred that metal salt of carboxylic acid he carbon numbers 6-8 contains. When using tetraethylene Glyco Lot's 2-ethylhexanoate neforth "4GO") as a plasticizer, it is preferred that metal salt of the carbon number 6 or boxylic acid of 7 contains.

[04]In order to prevent hydrolysis by heat at the time of film production of polyvinyl-acetal resin sticized (above-mentioned 3) as much as possible, it comprises with a plasticizer like triethylene ol di-heptanoate (3G7) or tetraethylene glycol di-heptanoate (4G7). It is preferred to use a side in type plasticizer like 3GH which cannot cause hydrolysis easily, 3GO, and 4GO, or a horse ckerel peat system type plasticizer like DHA.

[05]Above-mentioned 3GH has a track record long as a plasticizer for interlayers, and has the ntage of being hard to cause hydrolysis, as compared with 3G7 and 4G7 grade which an organic d ingredient of a raw material is a side chain type, and are straight chain types. For example as pared with 3GH, since the above-mentioned 3GO or 4GO are high boiling points, they have the ntage of being hard to vaporize at the time of film production or doubling processing.

[06]Above-mentioned 3GH, 3GO, 4GO, and DHA may use together with other plasticizers which y be used alone and mentioned later. Although a concomitant use rate in particular of above- oned 3GH, 3GO, 4GO and DHA, and other plasticizers is not limited, it is preferred that the unt of concomitant use of other plasticizers is less than 50% of the weight of a plasticizer of ve-mentioned 3GH, 3GO, 4GO, and DHA. Since the characteristic which above-mentioned 3GH, GO, 4GO, and DHA have can weaken with other plasticizers if it is 50 % of the weight or more, an et of an adhesive strength regulator used corresponding to these may not no longer be onstrated fully.

[07]Metal salt of carboxylic acid as an adhesive strength regulator used when setting a plasticizer he above-mentioned interlayer as a specific thing. Although not limited in particular, pentanoic d metal salt (carbon number 5), hexanoic acid (2-ethylbutanoic acid) metal salt (carbon number 6), tanoic acid metal salt (carbon number 7), octanoic acid metal salt (carbon number 8), etc. are oned, and those one sort or two sorts or more are suitably used according to a kind of the ve-mentioned plasticizer. The above-mentioned carboxylic acid may be a straight chain type, and y be a side chain type.

[08]The moisture resistance of an interlayer obtained may become insufficient in case of metal salt se carbon number of the above-mentioned carboxylic acid is too small, generating of althium y become large, and a fall preventive effect of adhesive strength of an interlayer and glass with passage of time may become insufficient conversely in case of metal salt whose carbon number arboxylic acid is too large.

[09]Metal salt of carboxylic acid as the above-mentioned adhesive strength regulator. Although it y be used independently, respectively, formic acid magnesium, magnesium acetate, A metal salt tem adhesive strength regulator of carboxylic acid of the carbon numbers 1-4 like magnesium

propionate and magnesium butanoate; it may be used together with other adhesive strength regulators, such as a denaturation silicone oil system adhesive strength regulator mentioned later. [010]When adding the above-mentioned alkali metal salt and/or alkaline earth metal salt as an adhesive strength regulator, the addition has 0.01 to 0.2 preferred weight section to polyvinyl-acetal resin 100 weight section. Since an adhesive strength adjustment effect will be lost if it is less than 0.01 weight sections, penetration-proof performance of a glass laminate obtained may fall. If 0.2 weight sections are exceeded, while spoiling the transparency of a glass laminate produced by carrying out bleed out, there is a possibility that adhesive strength of an interlayer and glass may decline too much. It is 0.03 to 0.09 weight section more preferably.

[011]As for sodium concentration, since especially a white bluish mark happens easily when the above-mentioned alkali metal salt is sodium salt, being set to 50 ppm or less is preferred. As for potassium element concentration, since a white bluish mark happens easily also when the above-mentioned alkali metal salt is potassium salt, being set to 100 ppm or less is preferred.

[012]The above-mentioned alkali metal salt and/or alkaline earth metal salt, Sulfuric acid used [ others / in a case of adding as the above-mentioned adhesive strength regulator ] in a reaction of polyvinyl-acetal resin. The above-mentioned metal salt may have contained in various raw materials, water, etc. which were used in a case where it originates in having used alkali metal salt and alkaline earth metal salt as a neutralizer of an acid catalyst of chloride etc., and a reaction of polyvinyl-acetal resin. Alkali metal salt and alkaline earth metal salt as the above-mentioned neutralizer can be diverted to some other purpose as an adhesive strength regulator.

[013]An interlayer for glass laminates of this invention consists of a plastic resin film which consists of additive agents, such as an above-mentioned dispersing agent and an adhesive strength regulator, by the above-mentioned polyvinyl-acetal resin, a plasticizer, and necessarily.

[014]As a plasticizer used in this invention, phosphoric acid system plasticizers, such as organic ester system plasticizers, such as a publicly known plasticizer used for this kind of interlayer, for example, monobasic acid ester, and polybasic acid ester, an organic phosphorus acid system, an organic phosphorus acid system, etc. are used.

[015]In the above-mentioned monobasic acid ester, for example Triethylene glycol, Glycol system ester obtained by a reaction with organic acid, such as butanoic acid, isobutyric acid, caproic acid, 2-ethylbutanoic acid, heptanoic acid, n-octylic acid, 2-ethylhexyl acid, pelargonic acid (n-nonylic acid), and decylacid, is preferred. In addition, ester of tetraethylene glycol, tripropylene glycol, and the above-mentioned organic acid is also used.

[016]As the above-mentioned polybasic acid ester, ester of organic acid, such as adipic acid, sebacic acid, and azelaic acid, and straight chain shape of the carbon numbers 4-8 or branched state alcohol is preferred, for example.

[017]As an example of the above-mentioned organic ester system plasticizer. Triethylene glycol di-2-ethyl butyrate, triethylene glycol di-2-ethylhexoate, Triethylene glycol dicaprylate, triethylene glycol di-n-octanoate, Triethylene glycol di-n-HEPULOETO, tetraethylene glycol di-n-HEPULOETO, other dibutyl sebacate, dioctylazelaate, and a dibutylcarbitol horse mackerel peat are used suitably.

[018]In addition, ethylene glycol di-2-ethyl butyrate, 1,3-propylene glycol di-2-ethyl butyrate, 1,4-pyrene glycol di-2-ethyl butyrate, 1,4-butyrene GLURKORUJ 2-ethyl butyrate, 1,2-butyrene-glycol di-2-ethylene butyrate, diethylene-glycol di-2-ethyl butyrate, Diethylene-glycol di-2-ethylhexoate, propylene glycol di-2-ethyl butyrate, Triethylene glycol di-2-ethylhexoate, tetraethylene glycol di-2-ethyl butyrate, diethylene-glycol JKAPURIEO, etc. are used as a plasticizer.

[019]As the above-mentioned phosphoric acid system plasticizer, triethoxyethyl phosphate, isodecyl phenyl phosphate, triisopropyl phosphate, etc. are preferred. It is preferred to blend a diester system compound which consists of dicarboxylic acid and monohydric alcohol, or consists of monocarboxylic acid and dihydric alcohol in the above-mentioned plasticizer.

[0120]As the above-mentioned amount of plasticizers, 20 to 70 weight section is preferred to polyvinyl-acetal resin 100 weight section, and it is 40 to 60 weight section more preferably. The penetration resistance of a glass laminate which will be obtained if it is less than 20 weight sections may fall, if 70 weight sections are exceeded, a plasticizer will carry out bleeding, optical strain may become large or the transparency of a resin layer and an adhesive property may fall.

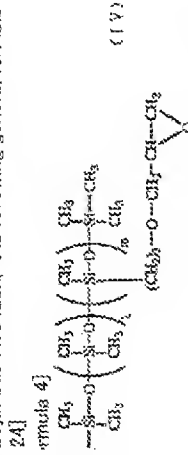
[0121]In this invention, a publicly known additive agent currently used for interlayers for glass laminates, such as conversion silicon oil for controlling penetration resistance besides a dispersing



ant and an adhesive strength regulator, an ultraviolet ray absorbent, light stabilizer, an antioxidant, surface-active agent, and colorant, as an additive agent can be blended.

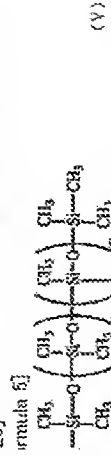
[22]An epoxy denaturation silicone oil which is not limited especially as the above-mentioned modified silicone oil, for example, is indicated by JP 55-29950, B. Ether denaturation silicone oil, ester denaturation silicone oil, and amine denaturation silicone oil carboxyl modified silicone oil etc. are mentioned. These modified silicone oil is fluids produced by making a compound which should naturalize to a polysiloxane resin generally.

[23]In this invention, it is following general formula (IV).

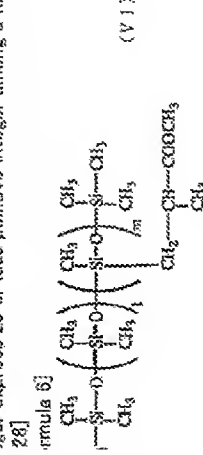


[25]Epoxy modified silicone oil, following general formula (V) which are expressed with (I) and m

express 30 or less positive integer among a formula)



integer express 20 or less positive integer among a formula), and following general formula (VI)



[29]Ester modified silicone oil expressed with (I and m express 30 or less positive integer among a formula) is used especially preferably. Although each modified silicone oil shown by the above-mentioned general formula (IV), (V), and (VI) is expressed with a structural formula of a block polymer, in this invention, what is expressed with a structural formula of a random copolymer is similarly.

[30]The above-mentioned modified silicone oil may be used alone, and may use two or more sorts ether. As for a molecular weight of the above-mentioned modified silicone oil, 800-5000 are preferred. If localization to the surface falls that it is less than 900 and 5000 is exceeded, compatibility with resin becomes poor, bleed out will be carried out, to a membrane surface, and adhesive strength with glass will fall to it. It is 1500-4000 more preferably.

[31]An addition of the above-mentioned modified silicone oil has 0.01 to 0.2 preferred weight ratio to the polyvinyl-acetal resin 100 above-mentioned weight section. If a preventive effect of the blush mark according that they are less than 0.01 weight sections to moisture absorption comes insufficient and exceeds 0.2 weight section, compatibility with resin becomes poor, bleed out will be carried out to a membrane surface, and adhesive strength with glass will fall to it. It is 0.03 to 0.1 weight section more preferably.

[32]It is not limited especially as the above-mentioned antioxidant, but as a thing of a phenol

system. For example, t-butylhydroxytoluene (BHT) ("Sumi Rider BHT (trade name)" by Sumitomo Chemical Co., Ltd.), Tetraakis [methylene-3-(3'-5'-di-t-butyl-4'-hydroxyphenyl) propionate] methane (RUCOA NOx 1010, Ciba-Geigy make) etc. are mentioned.

[113]It is not limited especially as the above-mentioned ultraviolet ray absorbent, but as a thing of a benzotriazol system, for example, 2-(2'-hydroxy-5'-methylphenyl) benzotriazol (the tinuvin P.). The Ciba-Geigy make, 2-(2'-hydroxy-3' and 5'-di-t-butylphenyl) benzotriazol (the tinuvin 320, Ciba-Geigy make), A 2-(2'-hydroxy-3'-t-butyl-5'-methylphenyl)-5-chlorobenzotriazole (the tinuvin 326, Ciba-Geigy make). A thing of hindered amine systems, such as tiling LA-57 (made by ADEKAGASU) of benzotriazol systems, such as 2-(2'-hydroxy-3' and 5'-di-aryl phenyl) benzotriazol (the tinuvin 328, Ciba-Geigy make), etc. are mentioned.

[114]As the above-mentioned light stabilizer, a thing of a hindered amine system, for example, "ADEKA stab LA-57" by the Asahi electricification company etc. (trade name) is mentioned. As the above-mentioned surface-active agent, sodium lauryl sulfate, alkylbenzene sulfonic acid, etc. are mentioned, for example.

[115]A manufacturing method of an interlayer for glass laminates of this invention. Although not limited in particular, other additive agents are blended with each above-mentioned resin [a plasticizer and if needed] for the specified quantity. A film is produced to a sheet shaped with an extrusion method, the calendar method, the pressing method, casting method, a tubular film process, etc., and a resin layer is fabricated, and let this be an interlayer, after kneading this uniformly.

[116]Thickness of the whole interlayer for glass laminates of this invention --- as a glass laminate --- the minimum --- when required penetration resistance and weatherability are taken into consideration, generally the range of 0.3-1.6 mm is preferred like thickness in the usual interlayer for glass laminates practical.

[117]As a glass plate used for the above-mentioned glass laminate, not only an inorganic clear glass board but organic clear glass boards, such as a polycarbonate board and a polymethylmethacrylate board, can be used.

[118]Especially as a kind of the above-mentioned inorganic clear glass board, it is not what is limited. Various inorganic glass, such as float sheet glass, polished plate glass, figured glass, wire glass, sheet glass containing a line, heat ray absorption plate glass, and colored sheet glass, etc. may be mentioned, these may be used independently, and two or more sorts may be used together. An inorganic clear glass board and an organic clear glass board may be laminated. Thickness of glass should just be suitably chosen by use and is not restricted in particular by it.

[119]In order to manufacture a glass laminate of this invention, a process of the usual glass laminate is adopted. For example, between transparent glass plates of two sheets, pinch an interlayer which consists of a resin layer fabricated by an above-mentioned method, and putting this into a rubber bag and carrying out decompression sucking at about 70-110 \*\*, carry out preliminary adhesion and it ranks second. It is manufactured by performing this adhesion by a pressure of about ten to 15 kg/cm<sup>2</sup> at about 120-150 \*\* using a press, using autoclave.

[120]In a manufacturing method of a glass laminate, heat crimping may be carried out at temperature of 80-100 \*\* at the same time it, makes the above-mentioned interlayer which comes at least to produce a film polyvinyl butyral resin plasticized between glass plates of a couple intervene and carries out suction deaeration under decompression. Put a layered product of a glass plate / interlayer / glass plate into a rubber bag, and more specifically for example, in autoclave, -For [10 to 30 minutes] grade heat crimping is carried out by a pressure about temperature of about 50-100 \*\*, and 1 - 10 kg/cm<sup>2</sup> carrying out suction deaeration under decompression of an about [ 500--- 100mmHg ], and it carries out by performing deaeration and adhesion simultaneously.

[121]By limiting temperature when carrying out heat crimping to the range of 80-100 \*\* as mentioned above in this manufacturing method, and setting up suitably terms and conditions, such as a sticking-by-pressure pressure, sticking-by-pressure time, and a decompression degree when carrying out suction deaeration, by within the limits about the above, it can adjust so that it may fall within the fitness range of a request of adhesive strength of an interlayer and glass.

[122]Although an example is hung up over below and this invention is explained to it in more detail, this invention is not limited only to these examples. The "part" in an example means a "weight section."

43]adding the average degree of polymerization 1700, and the degree% of polyvinyl alcohol 275g of saponification of 98.9 mol to the preparation pure water 2890g of Example 1 (1) resin --- warming --- dissolved. Temperature control of the system of reaction was carried out to 12 \*\*, 35% of chloride catalysts [ 201 g of ] and the *n*-butylaldehyde 148g were added, this temperature was held, and the reactant was deposited. Then, hold the system of reaction at 45 \*\* for 3 hours, make a reaction mixture, superfluous water washes, and unreacted *n*-butylaldehyde is flushed. The sodium hydroxide solution which is a general-purpose neutralizer neutralized the chloride catalyst, and also white colored polyvinyl butyral resin was obtained through rinsing and desiccation for 2 hours with superfluous water. The vinyl acetate ingredient (degree of acetalization) of this resin was 65.0-mol %, and the vinyl acetate component was 1.1-mol %.

44][2] Manufacture above-mentioned polyvinyl-butylal-resin 100 weight section of an interlayer, or mixing triethylene-glycol-di-2-ethyl-butylate 40 weight section as a plasticizer and fully mixing out melt kneading of this with a roll mill, with the press-forming machine, press forming was carried out for 30 minutes, and 150 \*\* of 0.76-mm-thick interlayers were obtained. The result of measuring the particle diameter of the sodium salt in an interlayer, and potassium salt by using of the secondary ion image using a time-of-flight type secondary-ion-mass-spectroscopy (ToF-SIMS) device (product [ made by PHIEVANS ] TFS-2000 type). The particle diameter of 1 monomer and potassium salt of the particle diameter of the sodium salt in an interlayer was less than 0.5 micrometer. The bottom was a result and 8 ppm in fixed quantity in ICP luminescence measurement analysis about the sodium element content of this interlayer. After ICP luminescence measurement analysis heats and decomposes a sample with sulfuric acid and nitric acid and sets the name of this decomposition product with ultrapure water, it is the method of quantifying by the AES method using an ICP-AES device (the "ICAP-575 type" by a Japanese jar rel ash company).

45][3] It inserts from both sides using the manufacture above-mentioned interlayer of a glass laminate with a transparent float glass (30 cm by 30 cmx2.5 mm in thickness). After putting this in rubber bag and carrying out indirect desulfurization mind with the degree of vacuum of 200torr for 30 minutes, it moved to 90 \*\* oven, deaerated, and the vacuum press was carried out, holding for 30 minutes at 80 more \*\*. Thus, this adhesion was performed for the glass laminate by which preliminary adhesion was carried out for 20 minutes on pressure 12 kg/cm<sup>2</sup> and conditions with a temperature of 1 \*\* in air type autoclave, and the transparent glass laminate was obtained. About the obtained glass laminate, an adhesive examination (pen Menu examination) and the moisture-proof white: bluish-ink sex test were done by the following methods.

46][A quality assessment (1) adhesive-property (pen Menu) examination glass laminate is neglected [ adjusted to -18 \*\*\*0.8 \*\* temperature, and this is struck with the hammer whose weight of a id is 0.45 kg, and it grinds until the particle diameter of glass is set to 6 mm or less. And the degree of exposure of the film after glass carries out partial avulsion is judged with the boundary sample which carried out grade attachment beforehand, as shown in Table 1. This is for the adhesive strength of a glass plate and an interlayer to judge whether it is in the predetermined range.

47]

ble 1]

剥離の露出度 (%)	ハンメル数
1.00	6
2.00	1
3.00	2
4.00	3
5.00	4
6.00	5
7.00	6
8.00	7
9.00	8

[0148][2] The moisture-proof white: bluish-mark sex-test resin layer was cut into the size of 4x4 cm. it was immersed in the ion exchange water of ordinary temperature (23 \*\*, and Hayas of 24 hours after was measured using the integral equation turbidity meter (made by Tokyo Denshoku Co., Ltd.). The result was shown in Table 2.

[0149]In preparation of resin of example 2 Example 1, washing and a washing process after putting in a neutralizer were changed in 1.5 hours, and it carried out like Example 1 except it. In this case, as for the sodium element content in the obtained interlayer, the particle diameter of 3 micrometers and potassium salt of the particle diameter of 13 ppm and sodium salt was less than 0.5 micrometer.

[0150]In preparation of resin of example 3 Example 1, it carried out like Example 1 except having changed washing and a washing process after putting in a neutralizer in 2.5 hours. In this case, as for the sodium element content in the obtained interlayer, the particle diameter of 0.5 micrometer and potassium salt of the particle diameter of 3 ppm and sodium salt was less than 0.5 micrometer.

[0151]In preparation of resin of example 4 Example 1, it carried out like Example 1 except having changed washing and a washing process after putting in a neutralizer in 3.5 hours. In this case, as for the sodium element content in the obtained interlayer, the particle diameter of less than 0.5 and potassium salt of the particle diameter of 0.9 ppm and sodium salt was less than 0.5 micrometer.

[0152]In preparation of resin of comparative example 1 Example 1, it carried out like Example 1 except having changed washing and a washing process after putting in a neutralizer in 1 hour. In this case, the particle diameter of 17 ppm and sodium salt of the sodium element content in the obtained interlayer was 6 micrometers.

[0153]In preparation of resin of comparative example 2 Example 1, it carried out like Example 1 except having changed washing and a washing process after putting in a neutralizer in 0.5 hour. The particle diameter of 35 ppm and sodium salt of the sodium element content in the obtained interlayer was 13 micrometers. The evaluation result of the above Examples 1-4 and the comparative examples 1 and 2 was shown in Table 2.

[0154]

[Table 2]

		乾燥度				比較例			
		1	2	3	4	1	2	3	4
中	アセチン-ブチルアルコール	1	1	1	1	1	1	1	1
	アセチン-ブチルアルコール (ppm)	85.8	85.8	85.8	85.8	85.8	85.8	85.8	85.8
	平均粒子径 (nm)	1700	1700	1700	1700	1700	1700	1700	1700
間	セリウムイオン成分 (%)	1.1	1.1	1.1	1.1	1.1	1.1	1.1	1.1
	乾燥度 (乾燥度)	40	40	40	40	40	40	40	40
	ナトリウム濃度 (ppm)	6	13	8	8	8	8	8	8
液	ナトリウム濃度 (ppm)	1	3	0.5	0.5	0.5	0.5	0.5	0.5
	カリウム濃度 (ppm)	0.5	0.5	0.5	0.5	0.5	0.5	0.5	0.5
	カリウム濃度 (ppm)	8	8	8	8	8	8	8	8
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
	カリウム濃度 (ppm)	24	27	24	24	24	24	24	24
水溶液	カリウム濃度 (ppm)	24	27	24	24	24</			

plete, the water of an excess (it is 30 times the amount to resin) washes, and unreacted n-butyraldehyde is flushed. The potassium hydroxide solution which is a general-purpose neutralizer neutralized the chloride catalyst, and also white powdered polyvinyl butyral resin was obtained through rinsing and deaeration for 2 hours with superfluous water. The vinyl-acetal ingredient (degree of acetalization) of this resin was 65.0-mol %, and the vinyl acetate component was 1.1-mol %.

57(2) Manufacture above-mentioned polyvinyl-butylal-resin, 100 weight section of an interlayer, or mixing triethylene-glycol-di-2-ethyl-butylate 40 weight section as a plasticizer and fully mixing out melt kneading of this with a roll mill, with the press-forming machine, press forming was carried out for 30 minutes, and 150 \*\* of 0.78-mm-thick interlayers were obtained. The bottom was a silt and 2.3 ppm in fixed quantity in ICP luminescence ultimate analysis about the potassium content of this interlayer. The particle diameter of less than [ 0.5 micrometer ] mum and potassium salt of the particle diameter of the sodium salt in an interlayer was less than 3 micrometers.

58(3) It inserts from both sides using the manufacture above-mentioned interlayer of a glass laminate with a transparent float glass (30 cm by 30 cmx2.5 mm in thickness). After putting this in in rubber bag and carrying out indirect desulfurization mind with the degree of vacuum of 200torr for minutes, it moved to 90 \*\* oven, deaerated, and the vacuum press was carried out, holding for 30 minutes at 80 more \*\*. Thus, this adhesion was performed for the glass laminate by which preliminary adhesion was carried out for 20 minutes on pressure 12 kg/cm<sup>2</sup> and conditions with a temperature of : \*\* in air type autoclave, and the transparent glass laminate was obtained.

59(3) Preparation of resin of example 6 Example 5, it carried out like Example 5 except having aged washing and a washing process after putting in a neutralizer in 2.5 hours. In this case, as for potassium elemental content in the obtained interlayer, the particle diameter of 1 micrometer and sum salt of the particle diameter of 5 ppm and potassium salt was less than 0.5 micrometer.

60(3) Preparation of resin of example 7 Example 5, it carried out like Example 5 except having aged washing and a washing process after putting in a neutralizer in 3.5 hours. In this case, as for potassium elemental content in the obtained interlayer, the particle diameter of less than 0.5 micrometer and sodium salt of the particle diameter of 0.7 ppm and potassium salt was less than 0.5 micrometer.

61(3) Preparation of resin of comparative example 3 Example 5, it carried out like Example 5 except having changed washing and a washing process after putting in a neutralizer in 1 hour. In this case, the particle diameter of 104 ppm and potassium salt of the potassium elemental content in the obtained interlayer was 6 micrometers.

62(3) Preparation of resin of comparative example 4 Example 5, it carried out like Example 5 except having changed washing and a washing process after putting in a neutralizer in 0.5 hour. In this case, the particle diameter of 220 ppm and potassium salt of the potassium elemental content in the obtained interlayer was 8 micrometers.

63(3) About the glass laminate obtained by the above Examples 5-8 and comparative examples 3 and 4, adhesive examination (pan Meru examination) and a humidity test are done by the same method as the above-mentioned, and the evaluation result is summarized in Table 3, and is shown.

64(3)

b6: 3(3)

	実施例	比較例			
		5	6	7	8
アセターナル塩基添加	4	4	4	4	4
アセターナル塩基濃度(%)	65.0	65.0	65.0	65.0	65.0
平均割合	1700	1700	1700	1700	1700
ビニルセターナル成分(%)	1.1	1.1	1.1	1.1	1.1
可溶性塩(%)	40	40	40	40	40
カリウム濃度(ppm)	22	5	0.7	104	220
カリウム塩(%)	3	1	0.5	5	8
ナトリウム濃度(ppm)	11.7	0.6	0.5	--	--
ナトリウム塩(%)	0.5	0.5	0.5	--	--
含有セターナルの割合(%)	8	8	8	8	7
水浸漬24時間後のヘイズ(%)	74	70	14	60	92
総合判定	○	○	○	×	×

[0165] The moisture resistance excellent in Examples 5-7 is shown so that clearly from the above-mentioned result.

[0166] Adding the average degree of polymerization 1700, and the degree of polyvinyl alcohol 275g of saponification of 98.9 mol to the preparation pure water 2890g of Example 8 (1) resin --- warming --- it dissolved. Temperature control of the system of reaction was carried out to 12 \*\*, 35% of chloride catalysts [ 201 g of ] and the n-butyraldehyde 149g were added, this temperature was held, and the reactant was deposited. Then, hold the system of reaction at 45 \*\* for 3 hours, make a reaction complete, superfluous water washes, and unreacted n-butyraldehyde is flushed. The sodium hydroxide solution which is a general-purpose neutralizer neutralized the chloride catalyst, and also white powdered polyvinyl butyral resin was obtained through rinsing and deaeration with superfluous water.

The degree of acetalization of this resin was 65.0-mol %.

[0167(2) Triethylene-glycol-di-2-ethyl-butylate 40 weight section was mixed with manufacture above-mentioned polyvinyl-butylal-resin 100 weight section of the interlayer as a plasticizer, press forming was carried out with the press-forming machine, and the interlayer was obtained. The bottom was a result and 13 ppm in fixed quantity in the ICP luminescence elemental analysis device about the sodium element content of this interlayer. The particle diameter of the sodium salt in an interlayer was 3 micrometers.

[0168(3) patching the manufacture above-mentioned interlayer of a glass laminate between the 2.5-mm-thick float glasses of two sheets, putting this in in a rubber bag, and carrying out suction deaeration under decompression of -600mmHg in autoclave. Heat crimping was simultaneously performed for 20 minutes on condition of the temperature of 80 \*\*, and pressure <sup>2</sup> of 5kg/cm, and the glass laminate was produced.

[0169] In production of the glass laminate of example 9 Example 8, the glass laminate was produced like Example 8 except the temperature at the time of heat crimping having been 80 \*\*. The particle diameter of the sodium salt in an interlayer was 3 micrometers.

[0170] In production of the glass laminate of example 10 Example 8, the glass laminate was produced like Example 8 except the temperature at the time of heat crimping having been 100 \*\*. The particle diameter of the sodium salt in an interlayer was 3 micrometers.

[0171] In preparation of resin of comparative example 5 Example 8, except having changed washing and a washing process for a short time a little, it carried out like Example 8 and the glass laminate was produced. In this case, the particle diameter of 30 ppm and sodium salt of the sodium element content in the obtained interlayer was 11 micrometers.

72]About the glass laminate obtained by the above Examples 8-10 and comparative example 5, adhesive examination (pan Meru examination) and the heat resistance test were done by the following methods. About the humidity test, it carried out like Example 1.

73]After cooling a valuation method (1) adhesive-property (pan Meru) examination glass laminate is 20 \*\* for 2 hours. An automatic hammer ring device is equipped, the whole surface of a glass laminate is uniformly hit by a hammer head, the area of the glass piece adhering to the interlayer of a laminate is observed visually, and adhesive strength (pan Meru value) is judged as contrasted to the grade sample shown in Table 1. The adhesive strength minimum is one point and the minimum of a grade sample is a ten-point method of ten points. Curved-surface shaping of the form is carried out with the curvature which is 50 mm in radius, the used automatic hammer ring is equipped with a hammer head with a blow effective diameter of 5 mm and a weight of 240g, and the striking power of a hammer head can be adjusted with a spring screw.

74]Take out, and observe the existence of foaming visually, after neglecting the obtained glass laminate under 130 \*\* atmosphere for 2 hours based on the heat resistance test JIS R3205 "glass laminate." The result of the above Examples 8-10 and the comparative example 5 was summarized in Table 4, and was shown.

75]

Table 4]

	実施例		比較例	
	8	9	10	5
トウモロ濃度(ppm)	13	13	13	30
トウモロ濃度(μm)	3	3	3	11
おせガラスの接着性 (パンメル値)	1	5	8	5
浸透24時間後のヘイズ (パンメル値)	37	38	33	58
おせガラスの耐熱性試験 の発泡の有無	なし	なし	なし	なし
合判定	○	○	○	×

76]adding the average degree of polymerization 1700, and the degree% of polyvinyl alcohol 275g of ionification of 98.9 mol to the preparation pure water 2890g of Example 11 (1) resin --- warning --- dissolved. Temperature control of the system of reaction was carried out to 12 \*\*, 35% of chloride catalysts { 201 g of } and the n-butylaldehyde 148g were added, this temperature was held, and the catalyst was deposited. Then, hold the system of reaction at 45 \*\* for 3 hours; the reaction was made to complete, the water of the excess (they are 30 times to resin) washed, and unreacted n-butylaldehyde was flushed. The pH value was 5.1 when pH of the system at this time was measured. Tied with the centering dehydrator and resin of water content was obtained 50%. White powdered vinyl butyral resin was obtained for this resin through the desiccation under the atmosphere of 60 mmHg and -700mmHg. The degree of acetalization of this resin was 65.0-mol %.

77]Triethylene-glycol-di-2-ethyl-butyrate 40 weight section was mixed with manufacture above-mentioned polyvinyl-butyral-resin 100 weight section of the interlayer as a plasticizer, pressing was carried out with the press-forming machine, and the interlayer was obtained. The bottom was a result and 0.7 ppm in fixed quantity in the ICP luminescence elemental analysis device about sodium element content in this interlayer. The particle diameter of the sodium salt in an interlayer is less than 0.5 micrometer.

78]pinching the manufacture above-mentioned interlayer of a glass laminate between the 2.5-mm-thick float glasses of two sheets, putting this in a rubber bag, and carrying out suction desiccation under decompression of -600mmHg in autoclave. Heat crimping was simultaneously

performed for 20 minutes on condition of the temperature of 80 \*\*, and pressure  $^2$  of 5kg/cm, and glass laminate was produced.

79]adding the average degree of polymerization 1700, and the degree% of polyvinyl alcohol 275g of

saponification of 98.9 mol to the preparation pure water 2890g of Example 12 (1) resin --- warning --- dissolved. Temperature control of the system of reaction was carried out to 12 \*\*, 35% of chloride catalysts { 201 g of } and the n-butylaldehyde 148g were added, this temperature was held, and the reactant was deposited. Then, hold the system of reaction at 45 \*\* for 3 hours, and the reaction was made to complete, the water of the excess (they are 30 times to resin) washed, unreacted n-butylaldehyde was flushed, and the chloride catalyst was neutralized using the sodium hydroxide solution which is a general-purpose neutralizer. Then, it rinsed with water with a temperature of 50 \*\* of an excess (they are 30 times to resin), and white powdered polyvinyl butyral resin was obtained through desiccation. The degree of acetalization of this resin was 65.0-mol %

[0180](2) Triethylene-glycol-di-2-ethyl-butyrate 40 weight section was mixed with manufacture above-mentioned polyvinyl-butyral-resin 100 weight section of the interlayer as a plasticizer, pressing was carried out with the press-forming machine, and the interlayer was obtained. The bottom was a result and 10 ppm in fixed quantity in the ICP luminescence elemental analysis device about the sodium element content in this interlayer. The particle diameter of the sodium salt in an interlayer was 3 micrometers.

[0181](3), pinching the manufacture above-mentioned interlayer of a glass laminate between the 2.5-mm-thick float glasses of two sheets, putting this in a rubber bag, and carrying out suction desiccation under decompression of -600mmHg in autoclave. Heat crimping was simultaneously performed for 20 minutes on condition of the temperature of 80 \*\*, and pressure 5 kg/cm $^2$ , and the glass laminate was produced.

[0182]In example 13 Example 12, the interlayer was obtained like Example 12 except [ all ] having used water with a temperature of 60 \*\* by washing after neutralization. The degree of acetalization of this interlayer was 65.0-mol %. The bottom was a result and 15 ppm in fixed quantity in the ICP luminescence elemental analysis device about the sodium element content in this interlayer. The particle diameter of the sodium salt in an interlayer was 4 micrometers.

[0183]adding the average degree of polymerization 1700, and the degree% of polyvinyl alcohol 275g of saponification of 98.9 mol to the preparation pure water 2890g of Example 14 (1) resin --- warning --- dissolved. Temperature control of the system of reaction was carried out to 12 \*\*, 35% of chloride catalysts { 201 g of } and the n-butylaldehyde 148g were added, this temperature was held, and the reactant was deposited. Then, hold the system of reaction at 45 \*\* for 3 hours, make a reaction complete, superfluous water washes, and unreacted n-butylaldehyde is flushed. Magnesium hydroxide which is a general-purpose neutralizer neutralized the chloride catalyst, and also it rinsed with superfluous water, and white powdered polyvinyl butyral resin was obtained through desiccation.

[0184](2) Triethylene-glycol-di-2-ethyl-butyrate 40 weight section of the interlayer was mixed with manufacture above-mentioned polyvinyl-butyral-resin 100 weight section of the interlayer as a plasticizer, pressing was carried out with the press-forming machine, and the interlayer was obtained. The bottom was a result and 0.7 ppm in fixed quantity in the ICP luminescence elemental analysis device about the sodium element content in this interlayer. The particle diameter of the sodium salt in an interlayer was less than 0.5 micrometer.

[0185](3), pinching the manufacture above-mentioned interlayer of a glass laminate between the 2.5-mm-thick float glasses of two sheets, putting this in a rubber bag, and carrying out suction desiccation under decompression of -600mmHg in autoclave. Heat crimping was simultaneously performed for 20 minutes on condition of the temperature of 80 \*\*, and pressure  $^2$  of 5kg/cm, and the glass laminate was produced.

[0186]About the glass laminate obtained in above Examples 11-14, the heat resistance test was done by the following methods. About the humidity test, it carried out like Example 1. However, Example 14 carried out only the humidity test.

[0187]The resin 1g was put into the valuation method (1) heat resistance test usual test tube, it heated in a 150 \*\* oil bath for 60 minutes, and the existence of degradation of resin was observed. The result of the above Examples 11-14 was summarized in Table 5, and was shown.

[0188]

[Table 5]







ガラス丸 粉砕物 (重量部)	ガラス丸 粉砕物 (重量部)	固体分含有割合		乾燥シリコ ンオイル (重量部)	水溶性エポキシ 樹脂 (重量部)
		重量	体積		
24	100	40	40/100	0.05	0.04
25	100	40	40/100	0.08	0.05
26	100	40	40/100	1.0	0.04
27	100	40	40/100	0.03	0.04
28	100	40	40/100	0.3	0.04
3	100	40	40/100	—	—
9	100	40	40/100	0.1	—

18]adding the average degree of polymerization 1700, and the degree% of polyvinyl alcohol 275g of  
-onification of 98.9 mol to the preparation pure water 2890g of example 29(1) polyvinyl-acetal resin  
warming — It dissolved. Temperature control of the system of reaction was carried out to 12 \*\*,  
g of 35-% of the weight chloride catalysts and the n-butylaldehyde 148g were added, this  
perature was held, and the reactant was deposited. With superfluous water, held the system of  
ction at 45 \*\* for 3 hours, make a reaction complete, wash. And Then, unreacted n-butylaldehyde  
hout. The sodium hydroxide solution which is a general-purpose neutralizer neutralized the  
oxide catalyst, and also white powdered polyvinyl butyral resin was obtained through rinsing and  
ication for 2 hours with superfluous water. The amount of 64-mol % and survival acetyl groups of  
degree of formation of average butyral of this resin was 1-mol %.

19](2) manufacture profit \*\*\* polyvinyl butyral resin (average-degree-of-polymerization 1700  
degree % of 64 mol of formation of average butyral) of the interlayer for glass laminates To  
out of residual acetyl groups 1 mol %100 copy, as a plasticizer 40 copies of triethylene glycol di-  
butyrate. And 0.13 copy of dimethyl octylamine is added as 0.75 copy of

ethylbenzenesulfonic acid, and amine as organic acid. After carrying out melt kneading enough with  
all mill, press forming was carried out for 30 minutes at 150 \*\* using the press-forming machine,  
the interlayer for glass laminates of 0.75 mm of average thickness was obtained. It was 50 ppm  
in the sodium content of the obtained interlayer was measured by ICP emission spectrometry. It  
s 4 micrometers when the particle diameter of the sodium element which exists in an interlayer  
s measured using the time-of-flight type secondary-ion-mass-spectrometry device (TOF-SIMS)  
20](3) It inserts from both slides using the manufacture above-mentioned interlayer of a glass  
inate with a transparent float glass (30 cm by 30 cmx3 mm in thickness). After putting this in in  
rubber bag and carrying out indirect desulfurization mind with the degree of vacuum of 200orr for  
minutes, it moved to 90 \*\* oven, deaerated, and the vacuum press was carried out, holding for 30  
utes at 80 more \*\*. Thus, this adhesion was performed for the glass laminate by which preliminary  
ession was carried out for 20 minutes on condition of the temperature of 135 \*\*, and pressure 12  
cm<sup>2</sup> in air type autoclave, and the glass laminate was obtained. About the obtained glass laminate  
rayer, the humidity test was done like Example 1 and the result was shown in Table 8.

21]In manufacture of the interlayer for example 30 glass laminates, 0.30 copy of octanoic acid is  
ed instead of 0.75 copy of dodecylbenzenesulfonic acid as organic acid. Except having added 0.35  
y of decyl amine instead of 0.13 copy of dimethyl octylamine as amine, the resin layer was  
ained like Example 29 and the evaluated result was shown in Table 9. It was 5 micrometers when  
particle diameter of the sodium element which exists in the obtained interlayer was measured like  
mple 29.

22]In manufacture of the interlayer for example 31 glass laminates, except having added 0.20 copy  
ik(2-ethylhexyl) phosphoric acid instead of 0.75 copy of dodecylbenzenesulfonic acid as organic  
d, the resin layer was obtained like Example 29 and the evaluated result was shown in Table 9. It  
s 2 micrometers when the particle diameter of the sodium element which exists in the obtained  
rayer was measured like Example 29.

23]In manufacture of the interlayer for comparative example 10 glass laminates, except not having  
led the organic acid and amine of the dispersing agent, the resin layer was obtained like Example  
and the evaluated result was shown in Table 9. It was 20 micrometers when the particle diameter

of the sodium element which exists in the obtained interlayer was measured like Example 29.  
[0224]In manufacture of the interlayer for comparative example 11 glass laminates, except not having  
added the organic acid and amine of the dispersing agent, and having changed washing time from 2  
hours in 3 hours, the resin layer was obtained like Example 29 and the evaluated result was shown in  
Table 9. It was 30 ppm when the sodium content which exists in the obtained interlayer was  
measured like Example 29. It was 13 micrometers when the particle diameter of the sodium element  
was measured like Example 29.

[0225]

[Table 9]

	実施例				比較例
	20	30	31	10	11
アセター-小量塩基量	4	4	4	4	4
平均ブチラール化率 (モル%)	64.0	64.0	64.0	64.0	64.0
減圧アセチル基量 (モル%)	1.0	1.0	1.0	1.0	1.0
平均含有率	1700	1700	1700	1700	1700
可塑剤量 (重量部)	40	40	40	40	40
サトリウム酸量 (ppm)	50	50	50	50	50
サトリウム酸量 (μmol)	4	5	5	20	13
水浸透 2 時間後のヘイズ	28	28	20	92	72
総合評価	○	○	○	×	×

[0226]Example 32 composition / preparation (composition of polyvinyl butyral resin) ion-exchange-  
water 2900 weight section, It heated and dissolved in 95 \*\*, having supplied degree% of polyvinyl  
alcohol of saponification of 99.2 mol 198 weight section (4.5 mol of vinyl alcohol considerable amount)  
to the reactor with an agitating device, and agitating it with the average degree of polymerization  
1700. After cooled this solution at 30 \*\*, having added 35-% of the weight chloride 196 weight section  
(1.9 mol) and n-butylaldehyde 152 weight section (2.1 mol), lowering solution temperature  
subsequently to 2 \*\*, holding this temperature and polyvinyl butyral resin's depositing, temperature  
up of the solution temperature was carried out to 30 \*\*, and it was held for 5 hours. After holding,  
sodium bicarbonate 147 weight section (1.7 mol) was added, it neutralized, rinsing and desiccation  
were performed, and the degree% of polyvinyl butyral resin of butyral-izing of 65 mol was obtained. It  
was 50 ppm when the sodium content of the obtained polyvinyl butyral resin was measured by ICP  
emission spectrometry. The particle diameter of sodium salt was 12 micrometers.

[0227](Production of a resin layer) Polyvinyl-butylal-resin 100 obtained weight section, Triethylene-  
glycol-di-2-ethyl-butylate 40 weight section, p-toluenesulfonic-acid 0.43 weight section, And press  
forming of the kneaded material produced by supplying hexylamine 0.23 weight section to a roll mill,  
and kneading it was carried out for 10 minutes on condition of 150 \*\* and 120 kg/cm<sup>2</sup> with the  
press-forming machine, and the 0.8-mm-thick resin layer was obtained. The obtained resin layer was  
used and the moisture-proof white-blush-mark sex test was done like Example 1. The result was  
shown in Table 16.

[0228]Instead of example 33 hexylamine 0.23 weight section, the resin layer was obtained and  
evaluated like Example 32 except having used tetradecylamine 0.49 weight section. The result was  
shown in Table 16.

[0229]Instead of example 34 p-toluenesulfonic-acid 0.43 weight section, the resin layer was obtained  
and evaluated like Example 32 except having used dodecylbenzenesulfonic acid 0.75 weight section.  
The result was shown in Table 10.

[0230]The resin layer was obtained and evaluated like Example 32 instead of example 35 p-  
toluenesulfonic-acid 0.43 weight section and hexylamine 0.23 weight section except having used







sphoric acid 0.21 weight section and N,N-dimethyl octylamine 0.09 weight section. The result was shown in Table 13.

72) The resin layer was obtained and evaluated like Example 56 instead of example 62 JI (n-butyl) phosphoric acid 0.16 weight section and dodecyl amine 0.14 weight except having used

triethylphosphoric acid 0.17 weight section and dodecyl amine 0.13 weight section. The result was shown in Table 13.

73) The resin layer was obtained and evaluated like Example 56 instead of comparative example 23 (n-butyl) phosphoric acid 0.16 weight section and dodecyl amine 0.14 weight except having used no (n-dodecyl) sodium phosphate 0.30 weight section. The result was shown in Table 13.

74) The resin layer was obtained and evaluated like Example 56 instead of comparative example 24 (n-butyl) phosphoric acid 0.16 weight section and dodecyl amine 0.14 weight except having used triethylphosphoric acid 0.17 weight section and dodecyl amine 0.13 weight section. The result was shown in Table 13.

75) The resin layer was obtained and evaluated like Example 56 instead of comparative example 23 (n-butyl) phosphoric acid 0.16 weight section and dodecyl amine 0.14 weight except having used no (n-dodecyl) sodium phosphate 0.30 weight section. The result was shown in Table 13.

例	リン酸		アミン		乾燥後 4 時間 後の pH	乾燥後 4 時間 後の pH
	R <sup>1</sup>	R <sup>2</sup>	R <sup>1</sup>	R <sup>2</sup>		
50	n-ブチル	n-ブチル	n-ブチル	n-ブチル	1.6	1.6, 5
51	n-ブチル	n-ブチル	n-ブチル	n-ブチル	1.7	1.7, 3
52	n-ブチル	n-ブチル	n-ブチル	n-ブチル	1.8	1.8, 6
53	n-ブチル	n-ブチル	n-ブチル	n-ブチル	1.9	1.9, 8
54	n-ブチル	n-ブチル	n-ブチル	n-ブチル	2.0	2.0, 10
55	n-ブチル	n-ブチル	n-ブチル	n-ブチル	2.1	2.1, 12
56	n-ブチル	n-ブチル	n-ブチル	n-ブチル	2.2	2.2, 14
57	n-ブチル	n-ブチル	n-ブチル	n-ブチル	2.3	2.3, 16
58	n-ブチル	n-ブチル	n-ブチル	n-ブチル	2.4	2.4, 18
59	n-ブチル	n-ブチル	n-ブチル	n-ブチル	2.5	2.5, 20
60	n-ブチル	n-ブチル	n-ブチル	n-ブチル	2.6	2.6, 22
61	n-ブチル	n-ブチル	n-ブチル	n-ブチル	2.7	2.7, 24
62	n-ブチル	n-ブチル	n-ブチル	n-ブチル	2.8	2.8, 26
63	n-ブチル	n-ブチル	n-ブチル	n-ブチル	2.9	2.9, 28
64	n-ブチル	n-ブチル	n-ブチル	n-ブチル	3.0	3.0, 30

76) Adding the average degree of polymerization 1700, and the saponification degree % of polyvinyl alcohol 275g of 98.9 mol to the preparation pure water 2890g of example 63(1) polyvinyl-acetal resin warming — it dissolved. Temperature control of the system of reaction was carried out to 12 °C. The weight of chloride catalysts [ 201 g of ] and the n-butylaldehyde 148g were added. This temperature was held, and the reactant was deposited. Then, hold the system of reaction at 45 °C for 2 hours, make a reaction complete, superfluous water washes, and unreacted n-butylaldehyde is dried. The sodium hydroxide solution which is a neutralizer neutralized the chloride catalyst. Still re superfluous water performed rinsing and desiccation for 2 hours, and white powdered polyvinyl alcohol resin was obtained. As for the amount of residual acetyl groups, the content [ of the average degree of polymerization of the obtained polyvinyl butyral resin ] of neutralizing salt (NaOH) of the ideal vinyl alcohol component amount was [ 1700 and degree of butyral-izing ] 2 micrometers in particle diameter of 20 ppm and neutralizing salt in sodium concentration 34-mol% 1-mol% 65-mol%. 77) As opposed to 100 copies of polyvinyl butyral resin obtained by the manufacture above of interlayer for glass laminates. As a plasticizer, 40 copies of triethylene glycol di-2-ethyl butyrate (H), 0.071 copy (2.8x10<sup>-4</sup> mol) of magnesium 2-ethylbutanoate (carbon number 6), the ultraviolet absorber, and the antioxidant were added as metal salt (adhesive strength regulator) of butyric acid, and stirring mixing was carried out uniformly. The content of the organic acid in the mentioned 30H was 100 ppm. Subsequently, the small extrusion machine (made by a trade name "lab PURASUTO mill" Oriental energy machine company) was equipped with the T die, and on the extrusion temperature of 80-180 °C, and a metallic mold outlet temperature of 200 °C conditions, the mixture obtained above was extruded, the film was produced, and the interlayer for glass laminates about 0.5 mm thick was obtained.

78) After controlling the humidity of the interlayer for glass laminates obtained by the manufacture above of the glass laminate so that water content may be 0.4 to 0.5 % of the weight in air-conditioned room, it put between two float glasses (2.4 mm in thickness), and preliminary adhesion was carried out by the rolling method. Subsequently, actual adhesion of the layered product which preliminary adhesion was carried out was carried out on condition of the temperature of 130

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79) and pressure 13 kg/cm<sup>2</sup> in autoclave, and the glass laminate was obtained.

80) The following methods estimated this pan Meru value of the performance of the glass laminate obtained by the evaluation above. The method of the statement estimated moisture resistance in the Example 1. The result was as being shown in Table 14.

81) The heat struck the glass laminate which was neglected for 16 hours and carried out temperature control under pan Meru value=18±0.6 °C temperature with the hammer which is 0.45 kg, and it ground until the particle diameter of glass was set to 5 mm or less.

82) Following the judging standard which judges with the boundary sample which carried out grade attachment of the degree of exposure of the interlayer after glass carries out partial adhesion beforehand, and shows the result in Table 1 — as a pan Meru value — a table — the bottom, a pan Meru value — (b) primary stage and (\*\*) 50% — one month after [ - ] and (\*\*) 50% — it asked about three conditions two months after —. The adhesive strength of an interlayer and glass is also so large that the above-mentioned pan-mer value is large, and the adhesive strength of an interlayer and glass is also so small that a pan Meru value is small.

83) The interlayer for glass laminates and the glass laminate were obtained like Example 63 except having made metal salt of carboxylic acid as shown in Table 14 as Example 64 — a 69 adhesive-strength regulator contain.

84) In manufacture of the interlayer for comparative example 25 glass laminates, as metal salt of carboxylic acid, instead of 0.071 copy of magnesium 2-ethylbutanoate, the interlayer for glass laminates and the glass laminate were obtained like Example 63 except having added 0.04 copy (2.8x10<sup>-4</sup> mol) of magnesium acetate (carbon number 2).

85) The interlayer for glass laminates and the glass laminate were obtained like Example 63 except having made metal salt of carboxylic acid as shown in Table 14 as a comparative example 26 adhesive-strength regulator contain.

86) The performance of the glass laminate obtained by Examples 64-69 and the comparative examples 25 and 26 was evaluated like Example 63. The result was as being shown in Table 14.

87)

88)

例	電解質	カルボン酸の全量 (酸当量)	酸当量 (N/100 ml)	初期	最終	乾燥後 4 時間 後の pH	乾燥後 4 時間 後の pH
64	CH <sub>3</sub> COOH	0.071 (2.8)	0.071 (2.8)	5	5	5	5
65	CH <sub>3</sub> COOH	0.071 (2.8)	0.071 (2.8)	5	5	5	5
66	CH <sub>3</sub> COOH	0.071 (2.8)	0.071 (2.8)	5	5	5	5
67	CH <sub>3</sub> COOH	0.071 (2.8)	0.071 (2.8)	5	5	5	5
68	CH <sub>3</sub> COOH	0.071 (2.8)	0.071 (2.8)	5	5	5	5
69	CH <sub>3</sub> COOH	0.071 (2.8)	0.071 (2.8)	5	5	5	5
70	CH <sub>3</sub> COOH	0.071 (2.8)	0.071 (2.8)	5	5	5	5
71	CH <sub>3</sub> COOH	0.071 (2.8)	0.071 (2.8)	5	5	5	5
72	CH <sub>3</sub> COOH	0.071 (2.8)	0.071 (2.8)	5	5	5	5

89) The polyvinyl butyral resin (average-degree-of-polymerization 1650 and degree % of 67 mol of butyral-izing) which adjusted the combination presentation of the interlayer for example 70 glass laminates like the case of Example 65 as polyvinyl-acetal resin 32 mol of amount % and survival vinyl alcohol component amount % of residual acetyl groups of 1 mol. As the sodium content of 20 ppm, 100 copies of particle diameter of 2 micrometers of neutralizing salt, and a plasticizer, 38 copies of triethylene glycol di-2-ethylhexanoate (3GO). The interlayer for glass laminates was obtained like Example 63 except having considered it as 0.071 copy (2.8x10<sup>-4</sup> mol) of magnesium 2-ethylbutanoate (carbon number 6), the ultraviolet ray absorbent and the antioxidant as metal salt of carboxylic acid. The glass laminate was obtained like Example 63 using the interlayer for glass laminates obtained above.

90) The interlayer for glass laminates and the glass laminate were obtained like Example 70 except having made metal salt of carboxylic acid as shown in Table 15 as Example 71 and e 72 adhesive-

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[006] Instead of example 88 camphor sulfonic acid 0.056 weight section, the resin layer was obtained by evaluating like Example 86 except having used mesitylene sulfonic acid 0.056 weight section. The result was shown in Table 18.

[007] Instead of example 89 camphor sulfonic acid 0.056 weight section and N,N-dimethyl octylamine 0.11 weight section, the resin layer was obtained and evaluated like Example 86 except having used isobutylbenzenesulfonic acid 0.06 weight section and pyridine 0.02 weight section. The result was shown in Table 18.

[008] Instead of example 90 camphor sulfonic acid 0.056 weight section and N,N-dimethyl octylamine 0.11 weight section, the resin layer was obtained and evaluated like Example 86 except having used isobutylbenzenesulfonic acid 0.061 weight section and p-toluidine 0.039 weight section. The result was shown in Table 18.

[009] Instead of example 91 camphor sulfonic acid 0.056 weight section and N,N-dimethyl octylamine 0.11 weight section, the resin layer was obtained and evaluated like Example 86 except having used cyclohexane 2-acetoxy acid 0.045 weight section and dodecyl amine 0.104 weight section. The result was shown in Table 18.

[010] Instead of example 92 camphor sulfonic acid 0.056 weight section and N,N-dimethyl octylamine 0.11 weight section, the resin layer was obtained and evaluated like Example 86 except having used isobutylbenzenesulfonic acid 0.042 weight section and dodecyl amine 0.06 weight section. The result was shown in Table 18.

[011] Instead of comparative example 31 camphor sulfonic acid 0.056 weight section and N,N-dimethyl octylamine 0.044 weight section, the resin layer was obtained and evaluated like Example 86 except having used pyridine 0.1 weight section. The result was shown in Table 18.

[012] Instead of comparative example 32 camphor sulfonic acid 0.056 weight section and N,N-dimethyl octylamine 0.044 weight section, the resin layer was obtained and evaluated like Example 86 except having used salicylic acid 0.1 weight section. The result was shown in Table 18.

[013] Instead of comparative example 33 camphor sulfonic acid 0.056 weight section and N,N-dimethyl octylamine 0.044 weight section, the resin layer was obtained and evaluated like Example 86 except having used camphor sulfonic acid 0.1 weight section. The result was shown in Table 18.

[014] Instead of comparative example 34 camphor sulfonic acid 0.056 weight section and N,N-dimethyl octylamine 0.044 weight section, the resin layer was obtained and evaluated like Example 86 except having used pyridine 0.1 weight section. The result was shown in Table 18.

[015] Instead of comparative example 35 camphor sulfonic acid 0.056 weight section and N,N-dimethyl octylamine 0.044 weight section, the resin layer was obtained and evaluated like Example 86 except having used pyridine 0.1 weight section. The result was shown in Table 18.

例	樹脂	添加量	アミン	添加量	水浸漬 24 時間後のヘイズ (%)
88	樟腦スルホン酸	0.056	N,N-ジメチルオクチルアミン	0.044	25.7
87	メチルベンゼンスルホン酸	0.045	N,N-ジメチルオクチルアミン	0.057	29.0
88	メチルベンゼンスルホン酸	0.056	N,N-ジメチルオクチルアミン	0.044	20.4
89	イソブチルベンゼンスルホン酸	0.061	ピリジン	0.02	21.4
90	イソブチルベンゼンスルホン酸	0.061	p-トルエンジアミン	0.039	19.8
91	1,1'-ビス(4-tert-ブチルフェニル)エーテル	0.045	N,N-ジメチルオクチルアミン	0.104	26.8
92	サリチル酸	0.044	N,N-ジメチルオクチルアミン	0.06	26.1
93	—	—	ピリジン	0.1	46.2
94	ポリビニルアルコール	0.1	—	—	35.1
95	樟腦スルホン酸ナトリウム	—	—	0.1	66.2
96	塩化ビニルエーテル	—	—	0.1	54.7

[016] Polyvinyl-butyl-resin 100 weight section prepared in example 93 Example 42. Triethylene-ol-di-2-ethyl-butylate 40 weight section, octanoic acid 0.4 weight section. On the same conditions as Example 42, it kneaded, press forming of N,N-dimethyl octylamine 0.11 weight section [the 2-ethylbutanoic acid magnesium 0.037 weight section was carried out, and the 0.8-mm-thick

resin layer was obtained. About the obtained resin layer, the moisture-proof white-blush-mark sex test was done like Example 1.

[031] The friction test was done by the following method about the glass laminate obtained by fastening the above-mentioned resin layer between the glass plates (4x4 cm) of two sheets, and obtaining a glass laminate. The obtained result was shown in Table 19.

[0318] (Friction test) After the glass laminate was immersed in underwater [with a temperature of 80 ± 3 °C] for one week, it dried in oven with a temperature of 80 ± 3 °C for 4 hours. After repeating the process of this underwater immersion and oven drying 3 times, the existence of exfoliation of the interlayer in the circumference of a glass laminate was checked visually.

[0319] The result obtained by performing production and evaluation of a resin layer and a glass laminate like Example 93 was shown in Table 19 except having changed the quantity of example 94 N,N-dimethyl octylamine into 0.28 weight sections.

[0320] The result obtained by performing production and evaluation of a resin layer and a glass laminate like Example 93 was shown in Table 19 except having changed the quantity of example 95 octanoic acid into 0.1 weight sections, and having changed the quantity of N,N-dimethyl octylamine into 0.06 weight sections, respectively.

[0321] Change the quantity of example 98 octanoic acid into 0.2 weight sections, and the quantity of N,N-dimethyl octylamine is changed into 0.08 weight sections, respectively. The result obtained by performing production and evaluation of a resin layer and a glass laminate like Example 93 was shown in Table 19 except having used 2-ethylhexanoic acid magnesium 0.045 weight section instead of 2-ethylbutanoic acid magnesium 0.037 weight section.

[0322] Change the quantity of example 97 octanoic acid into 0.1 weight sections, and the quantity of N,N-dimethyl octylamine is changed into 0.06 weight sections, respectively. The result obtained by performing production and evaluation of a resin layer and a glass laminate like Example 93 was shown in Table 19 except having used 2-ethylhexanoic acid magnesium 0.045 weight section instead of 2-ethylbutanoic acid magnesium 0.037 weight section.

[0323] Except having changed the quantity of example 98 octanoic acid into 0.1 weight sections, and having used decyl amine 0.06 weight section and 2-ethylhexanoic acid magnesium 0.045 weight section instead of N,N-dimethyl octylamine 0.11 weight section and 2-ethylbutanoic acid magnesium 0.037 weight section. The result obtained by performing production and evaluation of a resin layer and a glass laminate like Example 93 was shown in Table 19.

[0324] Except having used di(2-ethylhexyl) phosphoric acid 0.03 weight section instead of example 99 octanoic acid 0.4 weight section, and having changed the quantity of N,N-dimethyl octylamine into 0.02 weight sections. The result obtained by performing production and evaluation of a resin layer and a glass laminate like Example 93 was shown in Table 19.

[0325] Using the glass laminate obtained by the comparative example 35 comparative example 12, the result obtained by doing only a friction test like Example 93 was looked like [Table 19] and was shown.

[0326] The result obtained by performing production and evaluation of a resin layer and a glass laminate like Example 93 was shown in Table 19 except not having used comparative example 36 octanoic acid and N,N-dimethyl octylamine.

[0327] The result obtained by performing production and evaluation of a resin layer and a glass laminate like Example 93 was shown in Table 19 except not having used comparative example 37 N,N-dimethyl octylamine.

[0328] The result obtained by performing production and evaluation of a resin layer and a glass laminate like Example 93 was shown in Table 19 except not having used comparative example 38 octanoic acid.

[0329] Comparative example 39 octanoic acid and N,N-dimethyl octylamine are not used. The result obtained by performing production and evaluation of a resin layer and a glass laminate like Example 93 was shown in Table 19 except having used 2-ethylhexanoic acid magnesium 0.045 weight section instead of 2-ethylbutanoic acid magnesium 0.037 weight section.

[0330] Instead of comparative example 40 octanoic acid 0.4 weight section, N,N-dimethyl octylamine 0.11 weight section, and 2-ethylbutanoic acid magnesium 0.037 weight section. The result obtained by performing production and evaluation of a resin layer and a glass laminate like Example 93 was shown in Table 19 except having used octanoic acid 0.2 weight section and 2-ethylhexanoic acid magnesium





51]The glass laminate of Examples 100-103 by this invention showed the outstanding moisture resistance so that clearly from Table 20. On the other hand, the glass laminates of the comparative examples 42 and 43 with the particle diameter of greater than 10 micrometers of the potassium element which exists in the interlayer for glass laminates had bad moisture resistance.

52]Adding the average degree of polymerization 1700, and the degree% of polyvinyl alcohol 275g of ionification of 98.9 mol to the preparation pure water 2890g of example 104(1) polyvinyl-acetal in -- warming -- it dissolved. Temperature control of the system of reaction was carried out to 12 35% of chloride catalysts [ 201 g of ] and the n-butylaldehyde 148g were added, this temperature is held, and the reactant was deposited. Then, hold the system of reaction at 45 ° for 3 hours, like a reaction complete, superfluous water washes, and unreacted n-butylaldehyde is flushed. The lithium hydroxide solution which is a general-purpose neutralizer neutralized the chloride catalyst, also white powdered polyvinyl butyral resin was obtained through rinsing and desiccation for 2 1rs with superfluous water. The amount of 64-mol % and survival acetyl groups of the degree of nation of average butyral of this resin was 1-mol %.

53]2) As opposed to 100 copies of manufacture above-mentioned polyvinyl butyral resin erage-degree-of-polymerization 1700 and amount % of 1 mol of degree % and survival acetyl ups of formation of average butyral of 64 mol) of the interlayer for glass laminates. As a sticizer, 40 copies of triethylene glycol di-2-ethyl butyrate. After having added 0.75 copy of lacybenzenesulfonic acid as organic acid, adding 0.13 copy of dimethyl octylamine as amine and y carrying out melt kneading with a roll mill, with the press-forming machine, press forming was ried out for 30 minutes, and 150 ° of interlayers for glass laminates of 0.76 mm of average kness were obtained. It was 50 ppm as a result of measuring the sodium content of the obtained rlayer by ICP luminescence ultimate analysis.

54]It was 4 micrometers as a result of measuring the particle diameter of the sodium element ch exists in the interlayer for glass laminates obtained above using time-of-flight type secondary mass spectroscopy (TOF-SIMS).

55]3) It inserts from both sides using the manufacture above-mentioned interlayer of a glass inate with a transparent float glass (30 cm by 30 cmx2.5 mm in thickness). After putting this in in rubber bag and carrying out indirect desulfurization mind with the degree of vacuum of 200rr for minutes, it moved to oven, desecrated, and the vacuum press was carried out, holding for 30 utes at 80 more °. Thus, this adhesion was performed for the glass laminate by which preliminary esion was carried out for 20 minutes on pressure 12 kg/cm<sup>2</sup> and conditions with a temperature of 1 ° in air type autoclave, and the glass laminate was obtained.

56]4) About the glass laminates obtained by the evaluation above, the humidity test was done by same method as Example 100. The result was as being shown in Table 21.

57]In manufacture of the interlayer for example 105 glass laminates, as organic acid, instead of 3 copy of dodecylbenzenesulfonic acid. The interlayer for glass laminates and the glass laminate e obtained like Example 104 except having added 0.30 copy of octanoic acid and having added 5 copy of decyl amine instead of 0.13 copy of dimethyl octylamine as amine. Sodium content was ppm.

58]It was 5 micrometers when the particle diameter of the sodium element which exists in the rlayer for glass laminates obtained above was measured like Example 104.

59]In manufacture of the interlayer for example 106 glass laminates, the interlayer for glass inates and the glass laminate were obtained like Example 104 instead of 0.75 copy of icycylbenzenesulfonic acid as organic acid except having added 0.20 copy of di(2-ethoxy)phoric acid. Sodium content was 40 ppm.

60]It was 2 micrometers when the particle diameter of the sodium element which exists in the rlayer for glass laminates obtained above was measured like Example 100.

61]In manufacture of the interlayer for example 107 glass laminates, the interlayer for glass inates and the glass laminate were obtained like Example 104 as an adhesives regulator except ing added 0.50 copy of sodium stearate. Sodium content was 280 ppm.

62]It was 7 micrometers when the particle diameter of the sodium element which exists in the rlayer for glass laminates obtained above was measured like Example 104.

63]In manufacture of the interlayer for comparative example 44 glass laminates, the interlayer for

glass laminates and the glass laminate were obtained like Example 104 except not having added the organic acid and amine of the dispersing agent. Sodium content was 51 ppm.

[0364]It was 20 micrometers when the particle diameter of the sodium element which exists in the interlayer for glass laminates obtained above was measured like Example 104.

[0365]In manufacture of the interlayer for comparative example 45 glass laminates, the interlayer for glass laminates and the glass laminate were obtained like Example 104 not having added the organic acid and amine of the dispersing agent, and except having changed washing time from 2 hours to 3 hours. Sodium content was 30 ppm.

[0366]It was 13 micrometers when the particle diameter of the sodium element which exists in the interlayer for glass laminates obtained above was measured like Example 104.

[0367]The humidity test of the glass laminate obtained by Examples 105-107 and the comparative examples 44 and 45 was done like Example 104. The result was as being shown in Table 21.

[0368]

[Table 21]

	実測値					公差値	
	104	105	106	107	44	45	46
アセターノール添加量	4	4	4	4	4	4	4
平均ブチラーガ化度 (mol %)	64.0	64.0	64.0	64.0	64.0	64.0	64.0
残存アセチル基 (mol %)	1.0	1.0	1.0	1.0	1.0	1.0	1.0
平均重合度	1700	1700	1700	1700	1700	1700	1700
所添加量 (g)	40	40	40	40	40	40	40
ナトリウム濃度 (ppm)	50	45	40	280	51	30	30
ナトリウム塩基 (ppm)	4	5	3	7	25	13	13
乾燥後の残存アセチル基 (mol %)	1.4	1.4	1.0	1.7	4.2	3.0	3.0
総合評価	○	○	○	○	○	×	×

[0369]The glass laminate of Examples 104-107 by this invention showed the outstanding moisture resistance so that clearly from Table 21. On the other hand, the glass laminate of the comparative examples 44 and 45 with the particle diameter of greater than 10 micrometers of the sodium element which exists in the interlayer for glass laminates had bad moisture resistance.

[0370]

[Effect of the invention]It consists of above-mentioned composition in this invention.

Therefore, without spoiling basic performance required for glass laminates, such as transparency, weatherability, an adhesive property, and penetration resistance, even when moreover placed into a humid atmosphere, the interlayer for glass laminates and glass laminate with few white bluish marks of a glass laminate edge part can be provided.

[Translation done.]

## NOTES \*

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## ATTEN AMENDMENT

ing date] May 13, Heisei 11

endment 1]

ment to be Amended] Specification

m(s) to be Amended] Claim

ethod of Amendment] Change

posed Amendment]

am(s)]

am 1] An interlayer for glass laminates when it is an interlayer for glass laminates which consists of plasticized polyvinyl polyvinylacetate resin films and said 0.3-0.8-mm-thick interlayer is immersed in water, wherein Hayes of 24 hours after is 50% or less.

am 2] The interlayer for glass laminates according to claim 1 whose particle diameter of sodium in an interlayer is 10 micrometers or less.

am 3] The interlayer for glass laminates according to claim 1 or 2 whose particle diameter of sodium salt in an interlayer is 5 micrometers or less.

am 4] The interlayer for glass laminates according to claim 1, 2, or 3 whose sodium concentration in interlayer is 50 ppm or less.

am 5] The interlayer for glass laminates according to claim 1 whose particle diameter of potassium in an interlayer is 10 micrometers or less.

am 6] The interlayer for glass laminates according to claim 1 or 5 whose particle diameter of potassium salt in an interlayer is 5 micrometers or less.

am 7] The interlayer for glass laminates according to claim 1, 5, or 6 whose potassium concentration in an interlayer is 100 ppm or less.

am 8] The interlayer for glass laminates according to claim 1, 2, 3, 4, 5, 6, or 7 which is a thing containing a compound which can form sodium salt and potassium salt, and a complex.

am 9] The interlayer for glass laminates according to claim 1, 2, 3, 4, 5, 6, or 7 which is a thing containing amine which is compatible in organic acid which is compatible in resin and a plasticizer.

am 10] The interlayer for glass laminates according to claim 1, 2, 3, 4, 5, 6, 7, 8, or 9 which is a thing containing at least one sort chosen from a group which consists of alkali metal salt and alkaline earth metal salt.

am 11] The interlayer for glass laminates according to claim 10 whose alkali metal salt is a thing having a particle diameter of 3 micrometers or less and whose alkaline earth metal salt is a thing having a particle diameter of 3 micrometers or less.

am 12] The interlayer for glass laminates according to claim 10 or 11 whose alkali metal salt is the alkali metal salt of organic acid of the carbon numbers 5-16 and whose alkaline earth metal salt is the alkaline earth metal salt of organic acid of the carbon numbers 5-16.

am 13] A glass laminate characterized by making the interlayer for glass laminates according to claim 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, or 12 come to intervene between glass of a couple at least.

am 14] An interlayer for glass laminates whose particle diameter of sodium salt in an interlayer is 10 micrometers or less, and whose potassium salt in an interlayer is 10 micrometers or less, and whose sodium concentration in an interlayer is 50 ppm or less, and whose potassium concentration in an interlayer is 100 ppm or less.

[Claim 15] An interlayer for glass laminates whose particle diameter of sodium salt in an interlayer is 10 micrometers or less, and whose potassium salt in an interlayer is 100 ppm or less.

[Claim 16] The interlayer for glass laminates according to claim 14 or 15 whose sodium concentration in an interlayer is 50 ppm or less.

[Claim 17] An interlayer for glass laminates whose sodium concentration in an interlayer is 100 ppm or less, and whose potassium concentration in an interlayer is 100 ppm or less.

[Claim 18] An interlayer for glass laminates whose particle diameter of potassium salt in an interlayer is 10 micrometers or less, and whose sodium concentration in an interlayer is 100 ppm or less.

[Claim 19] An interlayer for glass laminates whose potassium concentration in an interlayer is 100 ppm or less, and whose sodium concentration in an interlayer is 100 ppm or less.

[Claim 20] An interlayer for glass laminates whose potassium concentration in an interlayer is 100 ppm or less, and whose sodium concentration in an interlayer is 100 ppm or less.

[Claim 21] An interlayer for glass laminates whose potassium concentration in an interlayer is 100 ppm or less, and whose sodium concentration in an interlayer is 100 ppm or less.

[Claim 22] The interlayer for glass laminates according to claim 14, 15, 16, 17, 18, 19, 20, or 21 which is a thing containing a compound which can form sodium salt and potassium salt, and a complex.

[Claim 23] The interlayer for glass laminates according to claim 14, 15, 16, 17, 18, 19, 20, or 21 which is a thing containing amine which is compatible in organic acid which is compatible in resin and a plasticizer.

[Claim 24] The interlayer for glass laminates according to claim 14, 15, 16, 17, 18, 19, 20, 21, 22, or 23 which is a thing containing at least one sort chosen from a group which consists of alkali metal salt and alkaline earth metal salt.

[Claim 25] The interlayer for glass laminates according to claim 24 whose alkali metal salt is a thing having a particle diameter of 3 micrometers or less and whose alkaline earth metal salt is a thing having a particle diameter of 3 micrometers or less.

[Claim 26] The interlayer for glass laminates according to claim 24 or 25 whose alkali metal salt is the alkali metal salt of organic acid of the carbon numbers 5-16 and whose alkaline earth metal salt is the alkaline earth metal salt of organic acid of the carbon numbers 5-16.

[Claim 27] A glass laminate making claims 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24, and 25 or an interlayer for glass laminates given in 20 come to intervene between glass of a couple at least.

[Translation done.]